Atmos. Chem. Phys. Discuss., 4, S1381–S1392, 2004
www.atmos-chem-phys.org/acpd/4/S1381/

Atmospheric Chemistry and Physics Discussions

ACPD

4, S1381–S1392, 2004

Interactive Comment

Interactive comment on "Highly resolved global distribution of tropospheric NO_2 using GOME narrow swath mode data" *by* S. Beirle et al.

S. Beirle et al.

Received and published: 28 July 2004

We acknowledge the very in-depth remarks of Rev#2. The major reviewer comments a) and d) are dealt with in our general reply. We answer the remaining questions in the following.

Reviewer comment: b) an assessment of results suffers from unknown effects (impact of clouds)

Reply: We further analyzed the effect of pixel size and cloud influence, and it turned out that the results of Fig. 9 are not as unexpected as stated in the ACPD manuscript, as we explained in detail in our general reply (item 5). Generally, clouds shield the boundary layer, and mostly lead to underestimated VCDs. However, our general results, i.e. the spatial pattern found in the high resolution map, do not suffer from cloud effects.

Reviewer comment: c) the description of the retrieval method is too short, even taking into account that the method is published elsewhere.

Reply: We agree and extended the description of our retrieval.

Reviewer comment: e) the results concerning isolated spots (e.g. Istanbul, Mexico) are not convincing since the NSM GOME pixels are still too large to resolve the extent of a city plume.

Reply: Indeed we cannot resolve a city-plume. However, in our discussion on isolated spots we point out two aspects:

- the extent of these spots is quite small all over the world. The limiting factor is the resolution of the GOME NSM, not the transport of NO2! For its clarity of explanation, this is a new result and allows us to estimate an upper bound for the mean lifetime.

- Though we can not resolve the city-plume, we still can estimate its extent. Our revised analysis of the smearing effect indicates that the pollution plume for Mexico City is about 60 km. We think that this is not trivial, since we can make a (reasonable) statement on the plume extent that is lower than the actual GOME NSM resolution.

Reviewer comment: In order to sharpen the profile you may highlight scientific work such as the seasonal correction and the analysis of the smoothing effect.

Reply: We explain our deseasonalization method in more detail now and stress it as a new and successful method. The quantitative comparison of the ratios of the NSM/SSM maxima (in the context of Fig. 6) is revised since we now also consider the fact that the NSM backscan is slightly smaller than the SSM forescan.

Reviewer comment: A more general remark is to open the view for development outside the local science community, e.g. in your reference list. Try to remove the grey literature (Ph.D. thesis, diploma thesis) wherever possible and replace it by corresponding articles.

ACPD

4, S1381–S1392, 2004

Interactive Comment

Reply: Of course, we prefer to cite peer-reviewed articles. Unfortunately, such references are not available for some aspects (yet), and in detail descriptions of methods etc. are often explained only in PhD theses. To make these references available for the scientific community, we added web links to the cited PhD theses.

Reviewer comment: The entire last paragraph of the abstract is somewhat unclear. What you want to say is the following: You can calculate differences of NO2 VCDs derived from large pixels and smaller pixels. Then you will notice a difference which can be further analysed. A quantitative analysis of these differences (are you still comparing apples and apples?) can be used to study the smearing effect. This is important for e.g. budget studies but it is not per se important for SCIAMACHY and other sensors. There, you will only have the smaller pixels and the GOME results help to interpret but do not influence the SCIAMACHY/GOME-2/OMI retrieval, or do they ? It tells you what you can expect from sensors with better resolution but it has to my knowledge no impact on the retrieval method.

Reply: Our formulation was unclear. We clarified that our results are important for comparisons of GOME with its successors (rather than their measurements themselves).

Reviewer comment: Introduction: p 1668, l16: These studies clearly showed, that...

Change the relation. It is misleading to speak about different sources of NOx because the reader now expects a description of source processes followed by an explanation of chemical transformations. Instead, you may just say that you are able to identify industrial regions of the world, which is actually a trivial statement. This is already known from standard GOME data. So a more precise formulation of this paragraph is required.

Reply: The logical order of the introduction is meant to be

- NOx is an important trace gas

- general sources and their strengths are (roughly) known

ACPD

4, S1381–S1392, 2004

Interactive Comment

- but uncertainties are quite high, and questions remain

- satellite measurements provide additional, new and independent data, as has been demonstrated in several studies

- but GOME suffers from the large east-west extent of the ground pixels, and a better spatial resolution is of high importance.

We structured the introduction in this order.

Reviewer comment: p 1668, l8 "...of the total column ()."

Add at least a brief description how this is done. Tell the reader about the main (hidden) assumptions !

Reply: We extended the retrieval paragraph, describe our stratospheric estimation and state the assumptions made.

Reviewer comment: p 1668, l9 ...due to the degradation of the GOME instrument or the diffuser plate.

Is the diffuser plate not part of the instrument ? Too sloppy. Be more precise in your statements. The entire instrument ages but the diffuser is presumably the most affected part of the instrument and this will have consequences for the retrieval. Studies about the degradation of GOME have been already published (Tanzi, C.P., et al. Performance Degradation of GOME Polarization Monitoring, Adv. Space Res. 23, 1393-1396, 2000).

Are measurements of stratospheric species not affected by the degradation ? Again too sloppy.

Reply: We apologize that our statements were too short and misleading. The diffuser plate has proven to cause (time dependent) artificial spectral structures interfering with the NO2 cross-section (Richter and Wagner, 2001). To account for this, we use a fixed solar reference in our DOAS fit (Wenig et al., 2004). As a result, our analysis becomes more sensitive to degradation effects of the instrument. However, since these effects

ACPD

4, S1381–S1392, 2004

Interactive Comment

bias the retrieved columns in the reference sector as well as outside, their influence on the tropospheric VCD (i.e. the difference) is rather small. We added the Tanzi reference.

Reviewer comment: Your correction factor falls from heaven. It is just 2 and nobody knows why. What is the variance of correction factors derived by others ? Why is 2 then a representative value ? Is there a seasonal dependency, a latitudinal dependency ?

Reply: We extended the description of our method. Our correction factor of 2 corresponds to a tropospheric AMF of 1 as published by Richter and Burrows (2002) and is a good representative estimate for not too high solar zenith angles ($<\tilde{}70^{\circ}$). We point out that we used this simple approach of a constant correction factor to avoid uncertainties arising from external data. For instance, if we would assume that the profile height is lower over polluted regions and take this information from a model, our results would not be independent any more.

As a consequence of our simple approach, our derived absolute numbers can deviate systematically for some regions. A seasonal dependency of the albedo may cause an artificial Şseasonal effectŤ, but is accounted for by our deseasonalization procedure.

However, the main focus of our study is the spatial distribution of tropospheric NO2 in high resolution, that is not undermined by our simplified assumptions, but instead we received an independent data product.

Reviewer comment: Also the discussion of cloud effects is too short. You say you donŠt tackle the cloud problem because it is not fully understood. In other words, since clouds are present almost everywhere, your paper is questionable in general because the impact of clouds is not known. The way out is either a) to say that you concentrate on cloud-free scenes (or scenes with a cloudiness below a certain threshold) or donŠt you ? I guess you do because the trop. NO2 below clouds cannot be seen by GOME and all your hot spots are then not visible. b) to discuss the calculation of VCDs in case of partially cloudy scene here rather than in chapter 5 where the reader is actually

ACPD

4, S1381–S1392, 2004

surprised to be confronted with a basic discussion how the VCD is calculated. This needs to be done earlier. I therefore suggest to add chapter 5 to chapter 2 which is (too) short anyway.

Reply: We extended the discussion of the general effect of clouds, i.e. shielding of the boundary layer, in chapter 2, and added a reference to chapter 5, where we discuss the different patterns of cloud fraction and their influence for SSM and NSM observations.

In this study, we actually do not discard clouded pixels, since the number of NSM observations is rather low, and the improvement of selecting only cloud free pixels would be outbalanced by the loss of data (Please note that for our deseasonalization method we need a full set of 3 forescans plus backscan. If the cloud threshold would be exceeded for one subpixel, the whole set of observations would have to be discarded!) However, Fig. 3c and Fig. 5 clearly demonstrate that we in spite of using also cloudy pixels, we can detect hot spots, and retrieve high gradients in the spatial distribution of tropospheric NO2.

Reviewer comment: Move section 2.2 up to section 2.1 and vice versa. The technical details about GOME could even be in a chapter of its own, e.g. "2 GOME instrument". Or you may omit section 2.2 here but referring to existing papers where GOME is described in detail, which is acceptable, as long as instrumental details are not vital for your retrieval.

Reply: We thank the reviewer for that suggestion. The crucial argument for not discarding cloudy pixels is that the number of NSM measurements is quite low. Therefore we followed the reviewers suggestion and swap the corresponding paragraphs. The re-ordered retrieval section starts with a short introductory paragraph summarizing the general features of GOME. Section 2.1 discusses the special NSM features, section 2.2 explains the retrieval of tropospheric NO2 VCDs.

Reviewer comment: Second paragraph: Your statement about "many more details" in Fig. 3b with respect to Fig. 3a is actually unsustainable. It is simply the limited space

ACPD

4, S1381–S1392, 2004

Interactive Comment

for images which makes it difficult to see the advantages of NSM pixels. Both images look very similar (even 3c, which is not mentioned here) and all feature in 3a are in 3b,c but there arenŠt prominent features in 3b,c which are not in 3a. So these images are not really appropriate to underline your statements.

Reply: Fig. 3 as published on ACPD is indeed inappropriate for illustrating the improvements of the NSM. However, it was originally formatted to fill a full page in portrait format (as well as Fig. 5 and Fig. 7). We will take care of the actual size of Figs. 3,5,7 in the final ACP version.

Reviewer comment: Why is the scatter of results in the SAA higher for smaller pixels (Fig. 3a versus Fig 3c)?

Reply: This is due to the number of measurements per grid pixel that are about 5 for the southern Atlantic (that is only seldom sampled in the NSM, and part of the measurements disturbed by the SAA are removed by a fit-quality check, see also Fig. 2), in contrast to about 500 SSM observations.

Reviewer comment: You mention biomass burning areas but the given example (Fig 4) is not convincing. I actually expected spots over Central Africa, Brazil, probably also Indonesia but the scatter over Brazil makes it difficult to distinguish between effects from uncorrected level 1 data and "true" biomass burning effects. In your plots (Fig 3a-c, Fig 4) NO2 values over Africa around and below the equator are generally high but including the Namib desert as well.

Reply: Fig. 3 (and the related discussion) was not intended to prove that the enhanced VCDs over Central Africa are due to biomass burning. To do this, monthly means of SSM GOME observations are appropriate, where we can find yearly maxima in June and July for the Congo region. But here we argue that the arising stripes are due to temporally inhomogeneous sampling. We illustrate this effect for Central Africa, where the seasonal cycle is strongest due to biomass burning (whereas it also exist for other regions like Namibia). The scatter over Brasil is due to the SAA. We reformulated the

ACPD

4, S1381–S1392, 2004

Interactive Comment

corresponding paragraph.

Reviewer comment: Fig 4 shows a BBE but the example suffers from enhanced NO2 values of the same order over the Atlantic (southwards from your points A and B).

Reply: This is very probably due to outflow of NO2 produced during the biomass burning season as can be seen in monthly means of SSM tropospheric NO2.

Reviewer comment: p1670, l20ff: Your explanation on how the seasonal effect is finally corrected is again very short. It is no luxury to make use of equations. In general, the paper will benefit from at least some basic formulae, also in the chapter about the retrieval method.

Reply: We extended this paragraph and added a formula on how we actually deseasonalized the NSM data.

Reviewer comment: p 1673f section 4.2 Your quantitative analysis of SSW and NSM pixels is interesting and may indeed be used for a further evaluation of the NO2 regional or global distribution. Your discussion of Fig 7 could be more elaborated since it provides the impact of small versus large pixels on retrieved NO2 columns.

Reply: The general smearing effect is illustrated in Fig. 6. The results of Fig. 7 (dipolar structures over NOx sources) are thus quite expected. However, additional information is gained on the location of sources in highly polluted regions like the Po valley. Furthermore, Fig. 7 illustrates the limitations of the SSM observations and clearly indicates, for what regions the interpretation of common GOME pixels is critical, e.g. the western Alpine mountains or the North Sea between London and Rotterdam. We discuss both aspects in 4.2.

Reviewer comment: It does however not contain information about the impact of the "true" NO2 profile shape that is appropriate for larger and smaller ground pixels. Unfortunately, you donŠt provide any information about the NO2 profile shape and how it is taken into account in the retrieval. I therefore (have to) guess that the same profile

ACPD

4, S1381–S1392, 2004

Interactive Comment

shape is assumed for small-scale and large-scale pixels. This is an acceptable first order assumption (e.g. for routinely processed data) but it compromises a quantitative discussion of NO2 VCDs over "hot spots". In fact, you would have to use a profile with higher NO2 loading in the PBL for smaller pixels and vice versa, in order to reflect the enhanced loading of NO2 where it is produced (and its limited lifetime).

Reply: The reviewer points out a sophisticated detail that is indeed important for the determination of appropriate AMFs. However, to keep our study as elementary as possible, we do not consider possible systematic variations in the profile shape over hot spots (and state this in the retrieval paragraph). If we would use a-priori information on where the sources of NOx are, our retrieved NO2 distribution would not be an independent result any more.

Reviewer comment: Milan is the largest city in the Po valley but your statement about its responsibility for high NO2 values in the entire Po Valley is in contradiction with your previous estimation about the NO2 lifetime. Assuming 17 hours lifetime and having a wind speed of about 1 m/s (which is too low in fact) transports NO2 only Ÿ60 km away from Milan. The Po Valley is simply Italians industrial heart and industry and especially traffic (and also private combustion in the winter season) are the main sources of NO2 in the densely populated region. Torino and Padua are certainly other spots but with respect to the NO2 burden the Po Valley is more like the German Ruhr bassin and the bordering Netherlands.

Reply: Of course the high pollution of the Po valley is due to the general high population density and stems from several cities and industrial centers. Nevertheless, the NO2 burden is not at all homogeneous over the Po valley as seems to be the case in Fig. 5b (since the VCDs exceed the color-scale and structures are not resolved). Actually, Milan is the outstanding source in the Po valley (the VCD over Milan is more than twice that over Venice, see also Table 1). Here Fig. 7 provides additional information on the location of major sources within the Po valley, and we can clearly identify Torino as well as Padova/Venice as further source spots, which is impossible for the SSM data.

ACPD

4, S1381–S1392, 2004

Interactive Comment

Reviewer comment: Torino is obviously responsible for the overestimation of NO2 above the Western Alps (not Milan).

Reply: Concerning the GOME pixel orientation and size, both cities lead to an overestimation of the mean VCD westwards for the SSM.

Reviewer comment: Mexico is certainly a spot with an east-west extent $<$ 80 km but this is again a trivial result since it is just driven by the pixel resolution of GOME.

Reply: Our revised analysis of the smearing effect indicates that the pollution plume for Mexico City is about 60 km. We think that this is not a trivial deduction, since we can make a (reasonable) statement on the plume extent that is lower than the actual GOME NSM resolution.

Reviewer comment: p 1674, chapter 5: Due to the high cloud albedo, this effect is nonlinear:

What you presumably had in mind to say is about the nonlinear contribution of cloudy/cloud-free subpixels to the total reflected intensity. This is always non-linear provided that the albedo differs for both scenes, but the effect becomes more prominent if one (subpixel) scene is brighter than the other one. Move the equation below Fig. 8 to here and it becomes more clear. Fig 8: Remove parts of the description (equation, conclusions) from here and add it to the text.

Reply: We revised the corresponding paragraph as well as Fig. 8 (see also item 5 of our general reply). We clarified the nonlinear effects illustrated in Fig. 8 in the text.

Reviewer comment: HICRU database: Nobody outside the German community (probably even not outside your local science community) will a) know this Diploma thesis (presumably in German) b) may easily have access to it. Please provide more information about the database and remove this reference.

Reply: Unfortunately, so far there is no peer reviewed publication on HICRU, but M. Grzegorski is currently preparing a manuscript. We have added a conference ab-

ACPD

4, S1381–S1392, 2004

stract as reference. Furthermore, we briefly explain the main features of HICRU in the manuscript and discuss why we use it (and not other cloud products). For the considered polluted regions, it corresponds generally well to the results of FRESCO.

Reviewer comment: Even more, the given numbers of the fractional cover do not differ that much. What is the standard deviation of the cloud fractional cover (CFC) values and is it a statistically significant difference ?

Reply: Generally, the relative threshold of the CFC is a stronger criterion for the NSM. I.e., a scene with a single cloud of 30 km diameter would be categorized as cloud free in the SSM, but as cloudy in the NSM. Despite this bias, we still found more cloud free pixels in the NSM! (Please note, that the given numbers changed since we now consider only summertime observations for polluted regions of the northern hemisphere in paragraph 5). The difference of the NSM/SSM cloud distributions is highly significant, as we checked with a Wilcoxon rank sum test.

Reviewer comment: GOME products are provided with its own cloud product with known limitations. Why is this product not used and what are the differences of the CFC derived from the ESA GOME product and the HICRU cloud product ?

Reply: In his diploma thesis M. Grzegorski compared the results of several GOME cloud retrievals. The official ICFA cloud product uses an a-priori cloud height information that introduces large errors (e.g. Koelemeijer and Stammes, Validation of GOME cloud fractions, JGR, 1999). The FRESCO algorithm is better for most cases, but results in too high CFCs over deserts. The work of M. Grzegorski demonstrates that HICRU is a very stable and reliable algorithm, especially for the (crucial) retrieval of low CFCs, due to a sophisticated calculation of the ground albedo information.

Reviewer comment: p 1675, Conclusion.See again general comments at the beginning. In fact, the quasi-linear relation between NO2 VCDs derived from forward and backscan pixels is surprising but remains unexplained. Generally enhanced NO2 values above clouds over polluted areas seem to be unrealistic, even if vertical transport from the PBL to upper levels in cumulus-like clouds and lightning is taken into account. This result needs to be analysed further before it can be accepted as a comprehensible research result.

Reply: We understand the main open question now, and can explain the results of Fig. 9 without the need to assume unrealistically large amounts of NO2 above the clouds.

As a result of our study, we expect that successors of GOME with higher spatial resolution find almost the same mean VCDs as GOME, instead of higher ones as expected in the discussion of our previous version of Fig. 8.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 1665, 2004.

ACPD

4, S1381–S1392, 2004

Interactive Comment